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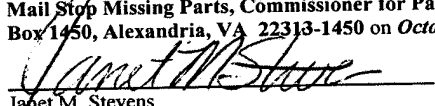
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For: MANUFACTURING METHOD OF PIEZOELECTRIC/ELECTROSTRICTIVE  
FILM TYPE DEVICE

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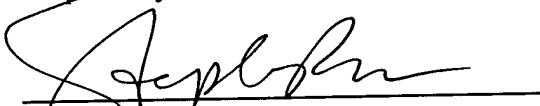
**TRANSMITTAL OF ENGLISH TRANSLATION  
OF NON-ENGLISH LANGUAGE *PROVISIONAL* APPLICATION  
UNDER 37 CFR §1.78(a)(5)**

Sir:

Applicants submit herewith an English translation of non-English language Provisional Application Serial No. 60/400,513, filed August 2, 2002 wherein the present application claims §119(e) priority of said non-English language provisional application (as noted in the Patent Application Bibliographic Data Sheet filed in the present application on July 30, 2003). Also submitted herewith is a statement by the translator that the translation is accurate.

Respectfully submitted,

October 29, 2003  
Date

  
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**NOTE: The attached translation is a translation of the Japanese-language *Provisional Application* from which priority is claimed in the present Non-Provisional Application.**



### VERIFICATION OF TRANSLATION

I, Koji Kikawa, state that I am fluent in the English language and in the Japanese language. I hereby verify that the attached English language translation of US Provisional Patent Application Serial No.60/400,513 entitled MANUFACTURING METHOD OF PIEZO ELECTRIC/ELECTROSTRICTIVE FILM TYPE DEVICE is a true and correct translation to the best of my knowledge and belief.

Signed this 22<sup>nd</sup> day October, 2003

By   
Koji Kikawa

TITLE OF THE INVENTION

MANUFACTURING METHOD OF PIEZOELECTRIC/ELECTROSTRICTIVE  
FILM TYPE DEVICE

5 BACKGROUND OF THE INVENTION AND RELATED ART

[0001] The present invention relates to a manufacturing method of a piezoelectric/electrostrictive film type device, and more particularly, to a manufacturing method of a piezoelectric/electrostrictive film type device, in which the piezoelectric/electrostrictive film type device having a high resonance frequency can be manufactured with a good efficiency while conduction of each electrode is secured.

10 [0002] In recent years, a piezoelectric/electrostrictive film type device has been used in various applications such as a displacement control device, solid device motor, ink jet head, relay, switch, shutter, pump, optical modulation device, and fin. The piezoelectric/electrostrictive film type device can control a minute displacement and moreover has superior properties such as a high electromechanical transduction efficiency, high-speed response, high durability, and low power consumption. In recent years, however, in the applications such as the ink jet head, from a demand for enhancement of printing quality and speed, there has been a demand for a device capable of a higher speed response.

25 [0003] By the way, in producing the piezoelectric/electrostrictive film type device, it is general to stack the lower electrode, a piezoelectric/electrostrictive layer, and

the upper electrode in order on a substrate of a ceramic;  
however, in order to secure insulation between the electrodes  
while avoiding dielectric breakdown of the  
piezoelectric/electrostrictive layer, as shown in FIG. 15, a  
5 piezoelectric/electrostrictive film type device 30 has been  
developed including a piezoelectric/electrostrictive layer 73  
disposed in such a size that the upper surface of the lower  
electrode 77 is coated and an end of the layer projects onto  
a substrate 44 (JP-A-6-260694 specification).

10 [0004] Moreover, in the conventional piezoelectric/  
electrostrictive film type device 30, a discontinuous plane  
is formed between a projecting portion 79 of the  
piezoelectric/electrostrictive layer 73 and the substrate 44,  
which is sometimes a cause for disconnection of the upper  
15 electrode 75; and therefore, it has also been disclosed that  
a gap between the projecting portion 79 of the piezoelectric/  
electrostrictive layer 73 and the substrate 44 is filled with  
a predetermined resin layer (said laid-open specification).

[0005] However, the piezoelectric/electrostrictive film  
20 type device provided with a resin layer, is very tiny, and it  
is difficult to coat only a predetermined portion with the  
resin layer. Therefore, the whole electrode has been coated  
with the resin layer under the present situation.

[0006] However, in the piezoelectric/electrostrictive film  
25 type device, a large number of devices are usually used in  
alignment with being electrically connected to one another.  
Therefore, it is quite important to manufacture the

electrodes of each device in such a state that the electrodes can be conducted to another device or external conducting means. Nevertheless, no consideration has been paid to this point in manufacturing the above-described piezoelectric/

5 electrostrictive film type device, and therefore, an operation of removing a part of the resin layer formed on each electrode after forming the resin layer has been required. Additionally, damages such as cracks are generated in the resin layer to be left at a removing time.

10 Furthermore, a part of the removed resin layer remains as dust in each portion of the device, and thus it has sometimes caused a trouble of incomplete contact at an electric property inspection time.

[0007] Moreover, in the conventional piezoelectric/  
15 electrostrictive device, based on recognition that flexural displacement or generating force is adversely influenced by the connection between the projecting portion of the piezoelectric/electrostrictive layer and the substrate, increase of rigidity of the device has not been considered at  
20 all; and therefore, a demand for achievement of a higher speed response in recent years has not necessarily sufficiently been satisfied.

#### SUMMARY OF THE INVENTION

25 [0008] The present invention has been developed in consideration of the above-described problem, and an object thereof is to provide a manufacturing method of a

piezoelectric/electrostrictive film type device, in which the piezoelectric/electrostrictive film type device having a high resonance frequency can efficiently be manufactured while securing conduction of each electrode.

5 [0009] That is, according to the present invention, there is provided a manufacturing method (This is hereinafter sometimes referred to as a "manufacturing method of a sandwiched piezoelectric/electrostrictive film type device".) of a piezoelectric/electrostrictive film type device  
10 including, a piezoelectric/electrostrictive operation portion containing a lower electrode, a piezoelectric/electrostrictive layer, and an upper electrode being stacked on a substrate of ceramic, wherein said method comprises the steps of:

15 forming the piezoelectric/electrostrictive layer of the piezoelectric/electrostrictive operation portion in a range broader than that of an applied portion of each electrode to project an end of the piezoelectric/electrostrictive layer;

20 coating a coating liquid prepared by admixing a polymerizable oligomer and inorganic particles in a dispersion medium in an amount sufficient to make the coating liquid permeable through a gap between at least a projecting portion of the piezoelectric/ electrostrictive layer and the  
25 substrate, and coat an each electrodes in such a position and an amount that does not coat at least a part of the electrode; and

drying the coating liquid to form a coupling member to couple the projected portion of the piezoelectric/electrostrictive layer to the substrate.

[0010] That is, according to the present invention, there

5 is provided a manufacturing method (This is sometimes hereinafter referred to as "a manufacturing method of a multilayered piezoelectric/electrostrictive film type device".) of a piezoelectric/electrostrictive film type device having a multilayered structure of the

10 piezoelectric/electrostrictive operation portion in which a plurality of electrodes and a plurality of piezoelectric/electrostrictive layers are alternately stacked on a substrate of ceramic, wherein said method comprises the steps of:

15 forming each piezoelectric/electrostrictive layers of the piezoelectric/electrostrictive operation portion in a range broader than that of an applied portion of each electrode to project an end of the piezoelectric/electrostrictive layer;

20 coating a coating liquid prepared by admixing a polymerizable oligomer and inorganic particles in a dispersion medium in an amount sufficient to make the coating liquid permeable through a gap between at least a projecting portion of the piezoelectric/electrostrictive layer and the  
25 substrate, and coat an each electrodes in such a position and an amount that does not coat at least a part of each electrode; and

drying the coating liquid to form a coupling member to couple the projected portion of the piezoelectric/electrostrictive layer to the substrate.

[0011] Here, in the specification, the term "a projected  
5 portion of the piezoelectric/electrostrictive layer" means a portion in which a lower or upper surface of the piezoelectric/electrostrictive layer does not contact an upper or a lower surface of each electrode. Moreover, the term electrode means that it includes, in addition to the  
10 applied portion brought in contact with the piezoelectric/electrostrictive layer, a terminal portion disposed to establish conduction to the electrode.

[0012] In the present invention, at the time of applying coating liquid, it is preferable to use a coating apparatus  
15 comprising: a pressurizing supply means for pressurizing/supplying a coating liquid in applying a coating liquid in a desired position and coat amount; a switching means disposed in a supply path of the pressurizing supply means to switch the supply of the coating liquid; and a discharge head for  
20 discharging the coating liquid introduced from the supply path of the pressurizing supply means to the outside, the discharge head comprising: a substrate including a coating liquid introduction path connected to the supply path of the pressurizing supply means, a pressurizing chamber in which  
25 the coating liquid introduction path opens, and one or more coating liquid discharge path(s) or discharge nozzle(s) communicating with and opening to the outside; and a



piezoelectric/electrostrictive operation portion disposed in a position corresponding to the pressurizing chamber on the substrate, and when the switching means is opened, the coating liquid introduced into the pressurizing chamber is continuously discharged in an atomized droplet state by flexural displacement of the piezoelectric/electrostrictive operation portion; or a coating apparatus comprising: a coating liquid introduction path connected to coating liquid supply source, a pressurizing chamber in which the coating liquid introduction path is opened, a substrate including one or more coating liquid discharge paths connected to the pressurizing chamber and opened to the outside, and a piezoelectric/electrostrictive operation portion disposed in a position corresponding to the pressurizing chamber on the substrate; and in accordance with flexural displacement of the piezoelectric/electrostrictive operation portion, the coating liquid introduced into the pressurizing chamber is discharged in an atomized droplet state.

[0013] Moreover, in the present invention, for the former

coating apparatus, a coating apparatus including a discharge head including a plurality of coating liquid discharge paths having different apertures, or a coating apparatus including a plurality of discharge heads having different apertures of the coating liquid discharge paths among the respective discharge heads is used; and for the latter apparatus, a coating apparatus including a plurality of coating liquid discharge paths having different apertures is preferably used

to preferably apply the coating liquid in an amount which differs depending on a discharge position. Moreover, the application of the coating liquid is preferably performed while vibrating at least the substrate or the  
5 piezoelectric/electrostrictive layer.

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a partially sectional view showing one embodiment of a sandwiched piezoelectric/electrostrictive  
10 film type device obtained by a manufacturing method of the present invention;

FIG. 2 is a partially sectional view showing another embodiment of the sandwiched piezoelectric/electrostrictive film type device obtained by the manufacturing method of the  
15 present invention;

FIG. 3 is a partially sectional view showing still another embodiment of the sandwiched piezoelectric/electrostrictive film type device obtained by the manufacturing method of the present invention;

FIG. 4 is a partially sectional view showing one embodiment of a multilayered piezoelectric/electrostrictive film type device obtained by the manufacturing method of the  
20 present invention;

FIG. 5 is a partially sectional view showing another  
25 embodiment of the multilayered piezoelectric/electrostrictive film type device obtained by the manufacturing method of the present invention;

FIG. 6 is a partially sectional view showing still another embodiment of the multilayered piezoelectric/electrostrictive film type device obtained by the manufacturing method of the present invention;

5           FIG. 7 is a top plan view schematically showing a state of the piezoelectric/electrostrictive film type device immediately after application of a different discharge amount of coating liquid in the manufacturing method of the present invention;

10           FIG. 8(a) is a top plan view of an on-demand type discharge apparatus for use in the manufacturing method of the present invention, and FIG. 8(b) is an A-A sectional view of FIG. 8(a);

15           FIG. 9(a) is a whole view schematically showing a continuous discharge apparatus for use in the manufacturing method of the present invention, and FIG. 9(b) is a partially enlarged sectional view showing a portion of a discharge head of FIG. 9(a);

FIG. 10 is a B-B sectional view of FIG. 8(a);

20           FIG. 11 is a top plan view schematically showing a state of the piezoelectric/electrostrictive film type device immediately after the application of the coating liquid having a different viscosity in the manufacturing method of the present invention;

25           FIG. 12 is a sectional view showing one example of a display device in which the piezoelectric/electrostrictive film type device obtained by the manufacturing method of the

present invention is used;

FIG. 13 is a sectional view showing one example of an ink jet printer head in which the piezoelectric/electrostrictive film type device obtained by the manufacturing method of the present invention is used;

FIG. 14(a) is a partially sectional view showing one embodiment of the multilayered piezoelectric/electrostrictive film type device including a loosely bonded layer obtained by the manufacturing method of the present invention;

FIG. 14(b) is a partially sectional view showing one embodiment of the multilayered piezoelectric/electrostrictive film type device in which micro-pores obtained by the manufacturing method of the present invention are filled; and

FIG. 15 is a partially sectional view showing one embodiment of a conventional piezoelectric/electrostrictive film type device.

#### DESCRIPTION OF PREFERRED EMBODIMENTS

[0014] Embodiments of a piezoelectric/electrostrictive film type device according to the present invention will concretely be described hereinafter, but the present invention is not limited to these when interpreted, and the present invention can variously be altered, modified, or improved based on knowledge of a person of ordinary as long as it does not deviate from the scope of the present invention.

[0015] A manufacturing method of the sandwiched

piezoelectric/electrostrictive film type device of the present invention is the one in which a piezoelectric/electrostrictive operation portion is arranged on a substrate of a ceramic by stacking a lower electrode, a piezoelectric/electrostrictive layer, and an upper electrode; and characterized by comprising the steps of forming the piezoelectric/electrostrictive layer of the piezoelectric/electrostrictive operation portion in a range broader than that of an applied portion of each electrode to project an end of the piezoelectric/electrostrictive layer, coating a coating liquid prepared by admixing a polymerizable oligomer and inorganic particles in a dispersion medium in an amount sufficient to make the coating liquid permeable through a gap between at least a projecting portion of the piezoelectric/electrostrictive layer and the substrate, and coat an each electrodes in such a position and an amount that does not coat at least a part of the electrodes; and drying the coating liquid to form a coupling member to couple ends of a projected portion of the piezoelectric/electrostrictive layer to the substrate. Moreover, the manufacturing method a multilayered piezoelectric/electrostrictive film type device of the present invention is the one provided with a piezoelectric/electrostrictive operation portion wherein a plurality of electrodes and a plurality of piezoelectric/electrostrictive layers are alternately stacked on a substrate of ceramic; characterized in that each piezoelectric/electrostrictive layer of the

piezoelectric/electrostrictive operation portion is disposed in a range broader than that the applied portion of each electrode with projecting an end of the piezoelectric/electrostrictive layer, coating a coating liquid prepared by admixing a polymerizable oligomer and inorganic particles in a dispersion medium in an amount sufficient to make the coating liquid permeable through a gap between at least a projecting portion of the piezoelectric/electrostrictive layer and the substrate, and coat an each electrodes in such a position and an amount that does not coat at least a part of the electrodes; and drying the coating liquid to form a coupling member to couple ends of a projected portion of the piezoelectric/electrostrictive layer to the substrate. By doing so, there can be manufactured a piezoelectric/electrostrictive film type device superior in a high speed response while retaining a flexural displacement equal to or more than that of a conventional piezoelectric/electrostrictive film type device in a secured state of conduction of each electrode without requiring an operation of removing a coated layer on the electrode. The present invention will concretely be described with reference to the drawings.

[0016] As shown in FIGS. 1 to 6, a substrate 44 for use in the present invention includes, for example, a structure in which a thin portion 66 having a thin plate shape is formed integrally with a fixing portion 68 formed of a thick ceramic member. Moreover, in the substrate 44 having this structure,

the thin portion 66 having a sheet-like shape is solidly attached to the fixing portion 68 in a position other than a position where a piezoelectric/electrostrictive layer 73 is disposed, and a cavity 48 is usually formed under the thin portion 66 so as to correspond to the position where the piezoelectric/electrostrictive layer 73 is disposed.

[0017] Moreover, as shown in FIGS. 3 and 6, the thin portion 66 may also be formed of a flat plate whose section of a thickness direction is rectangular. The thin portion 66 whose middle portion has a shape bent toward the cavity 48 as shown in FIGS. 2 and 5 or whose section of the thickness direction has a W shape as shown in FIGS. 1 and 4 is preferable in that the flexural displacement is large. The latter thin portion having a W-shaped section is especially preferable.

[0018] The thin portion 66 having the bent shape as shown in FIGS. 2 and 5 or the W shape as shown in FIGS. 1 and 4 can be formed by using shrinkage of a piezoelectric/electrostrictive film in the direction of width at a time of a sintering step of the piezoelectric/electrostrictive layer 73, or by adjusting a sintering shrinkage start timing or sintering shrinkage of upper and lower portions of the piezoelectric/electrostrictive layer 73, or further the shape of the thin portion 66.

[0019] In the present invention, the thickness of the thin portion 66 is preferably set to such a range that mechanical

strength of the device is secured and the flexural displacement of the piezoelectric/electrostrictive layer 73 is further prevented from being deteriorated by an increase of rigidity, and specifically, the thickness is in a range of preferably 1  $\mu\text{m}$  to 50  $\mu\text{m}$ , more preferably 3 to 50  $\mu\text{m}$ , especially preferably 3 to 12  $\mu\text{m}$ . The thickness of the fixing portion 68 is preferably 10  $\mu\text{m}$  or more, more preferably 50  $\mu\text{m}$  or more.

[0020] The shape of the surface of the substrate 44 on which a piezoelectric/electrostrictive operation portion 78 is mounted is not particularly limited to the rectangular shape, and the surface may also be formed in a circular shape, or a polygonal shape other than a square shape, such as a triangular shape.

[0021] The substrate 44 for use in the present invention may be formed of a ceramic material, but is preferably prepared by a material which does not change in properties at a time of a heating treatment of the piezoelectric/electrostrictive layer 73, the electrodes 75, 77, or the like, stacked on the thin portion 66 and which is superior in heat resistance and chemical stability. The substrate 44 is preferably formed of an electric insulation material so that a wiring connected to a lower electrode 77 formed on the substrate 44 is electrically disconnected.

[0022] Concretely, examples of the material include at least one ceramic material selected from a group consisting of (stabilized) zirconium oxide, aluminum oxide, magnesium



oxide, titanium oxide, cerium oxide, spinel, mullite, aluminum nitride, silicon nitride, and glass. Above all, the material containing stabilized zirconium oxide is preferable in that the mechanical strength is high, tenacity is superior, chemical stability is high, and reactivity with the piezoelectric/electrostrictive layer 73 or the electrodes 75, 77 is remarkably small.

[0023] Moreover, examples of stabilized zirconium oxide may contain stabilizers such as calcium oxide, magnesium oxide, yttrium oxide, scandium oxide, ytterbium oxide, cerium oxide, and rare earth metal oxide. For an added amount of the stabilizer, the amount of yttrium oxide or ytterbium oxide is preferably 1 to 30 mol%, more preferably 1.5 to 10 mol%. The amount of cerium oxide is preferably 6 to 50 mol%, more preferably 8 to 20 mol%. The amount of calcium oxide or magnesium oxide is preferably 5 to 40 mol%, more preferably 5 to 20 mol%.

[0024] Furthermore, among these stabilizers, yttrium oxide is more preferably added in an amount of preferably 1.5 to 10 mol%, particularly preferably 2 to 4 mol%.

[0025] Additionally, the thin portion 66 may contain, in addition to the ceramic described above, components such as silicon oxide or boron oxide contained in clay for use as a sintering aid. However, when these components are excessively contained, it is difficult to maintain a specific composition of the piezoelectric/ electrostrictive layer 73 by reaction between the substrate 44 and

piezoelectric/electrostrictive layer 73, and this is a cause for deterioration of piezoelectric/electrostrictive properties. Therefore, for the thin portion 66 of the substrate 44 in the present invention, silicon oxide or boron oxide contained in clay is in a range of preferably 20% or less by mass, more preferably 3% or less by mass in the material constituting the thin portion 66.

[0026] Furthermore, the ceramic constituting the thin portion 66 has an average particle diameter of crystal grains in a range of preferably 0.05 to 2  $\mu\text{m}$ , more preferably 0.1 to 1  $\mu\text{m}$  in order to enhance the mechanical strength of the thin portion 66.

[0027] Next, in the manufacturing method of the present invention, for a sandwiched piezoelectric/ electrostrictive film type device 10 shown in FIGS. 1 to 3, a piezoelectric/electrostrictive operation portion 78 in which the lower electrode 77, piezoelectric/electrostrictive layer 73, and upper electrode 75 are stacked is disposed on the substrate 44; and for a multilayered piezoelectric/electrostrictive film type device 20 shown in FIGS. 4 to 6, the piezoelectric/electrostrictive operation portion 78 in which a plurality of electrodes 75 to 77 are stacked alternately with a plurality of piezoelectric/electrostrictive layers 71, 72 is disposed on the substrate 44. Moreover, in the present invention, in any piezoelectric/electrostrictive film type device 10 (20), the piezoelectric/electrostrictive layer 73 (71, 72) of the piezoelectric/electrostrictive operation

portion 78 is formed in a range broader than that of the applied portion of each electrodes 75 to 77 (the lower electrode 77 and upper electrode 75 for the sandwiched piezoelectric/electrostrictive film type device), and an end of the piezoelectric/electrostrictive layer is projected.

[0028] By this, even in any piezoelectric/ electrostrictive film type device 10 (20), insulation of the respective electrodes 75, 77 (75 to 77) is firmly secured, and there can be provided the piezoelectric/electrostrictive film type device 10 (20) which is free of dielectric breakdown or short circuit. It is to be noted that in the present invention, for the piezoelectric/electrostrictive film type device 20 including the multilayered structure shown in FIGS. 4 to 6, the rigidity of the whole piezoelectric/electrostrictive operation portion 78 increases, and by a synergistic effect with a coupling member described later, there can be obtained the piezoelectric/electrostrictive film type device which has a high resonance frequency and in which a high speed response is possible; and in this respect, the electrodes 75, 77 are preferably disposed in the uppermost and the lowermost layers in the piezoelectric/ electrostrictive operation portion 78.

[0029] Moreover, the electrodes 75, 77 (75 to 77) are preferably formed of materials which are solid at room temperature and which can withstand a high-temperature oxidation atmosphere at a time of sintering and integrating the electrodes and the substrate and/or the piezoelectric/electrostrictive layer and which is superior in

electric conductivity. Specific examples of the materials include aluminum, titanium, chromium, iron, cobalt, nickel, copper, zinc, niobium, molybdenum, ruthenium, palladium, rhodium, silver, tin, tantalum, tungsten, iridium, platinum, gold, lead, and another simple metal or an alloy of these. A cermet material may also be used in which the material constituting the piezoelectric/electrostrictive layer, or the material constituting the substrate 44 such as zirconium oxide, cerium oxide, and titanium oxide is dispersed in the metals described above.

[0030] Moreover, it is preferred to choose a material for the electrodes 75, 77 (75 to 77) in the present invention, taking into consideration a method of forming the piezoelectric/electrostrictive layer 73 (71, 72). For example, in the manufacturing method of the sandwiched piezoelectric/electrostrictive film type device 10 shown in FIGS. 1 to 3, at a time of the heating treatment of the piezoelectric/electrostrictive layer 73, in the lower electrode 77 already formed on the substrate 44, it is preferable to use a simple metal of a platinum group which does not change even at a heating treatment temperature of the piezoelectric/electrostrictive layer 73, an alloy of the simple metal of the platinum group and gold and/or silver, an alloy of the platinum group metals, an alloy of two or more different types of metals of the platinum group, or high-melting metals such as an alloy of the metals of the platinum group and gold and/or silver. Also in the manufacturing

method of the multilayered piezoelectric/electrostrictive film type device 20 shown in FIGS. 4 to 6, it is preferable to use the high-melting metals in the electrode 77 positioned in the lowermost layer and an intermediate electrode 76

5 disposed between the piezoelectric/electrostrictive layers 71 and 72, which are already formed at the time of the heating treatment of the piezoelectric/electrostrictive layers 71, 72.

[0031] On the other hand, in the sandwiched

piezoelectric/electrostrictive film type device 10 shown in  
10 FIGS. 1 to 3, after the heating treatment of the

piezoelectric/electrostrictive layer 73, the upper electrode 75 can be formed on the piezoelectric/electrostrictive layer 73 at a low temperature. Alternatively, in the multilayered

piezoelectric/electrostrictive film type device 20 shown in

15 FIGS. 4 to 6, the electrode 75 positioned in the uppermost layer can be formed at the low temperature; and therefore, in addition to the high-melting metals, low-melting metals such as aluminum, gold, and silver may also be used.

[0032] Moreover, in the multilayered piezoelectric/

20 electrostrictive film type device shown in FIGS. 4 to 6, the electrode 77 positioned in the lowermost layer, and the intermediate electrode 76 disposed between the

piezoelectric/electrostrictive layers 71, 72 are also

preferably constituted of electrode materials which contain  
25 platinum as a major component and further contain additives

such as zirconium oxide, cerium oxide, and titanium oxide.

Although reasons are unclear, the electrodes and

piezoelectric/electrostrictive device constituted of these materials can be prevented from peeling. Moreover, the additives are preferably contained in all the electrode materials in an amount of 0.01 to 20% by mass in that a  
5 desired peel preventive effect is obtained.

[0033] In the present invention, examples of the method of forming the electrodes include ion beam, sputtering, vacuum deposition, PVD, ion plating, CVD, plating, screen printing, spraying, and dipping.

10 [0034] In the present invention, thicknesses of the electrodes 75, 77 (75 to 77) may be set to be appropriate in accordance with applications; and if they have excess thicknesses, the electrodes function as relaxing layers, and the flexural displacement is easily reduced, and therefore, a  
15 thickness of 15  $\mu\text{m}$  or less is preferable, and a thickness of 5  $\mu\text{m}$  or less is more preferable.

[0035] Next, in the present invention, in the case of the sandwiched piezoelectric/electrostrictive film type device shown in FIGS. 1 to 3, the piezoelectric/electrostrictive  
20 layer 73 (71, 72) can be obtained by the heating treatment at a predetermined temperature after applying a predetermined piezoelectric/electrostrictive material on the electrodes 75, 77. Moreover, in the case of the multilayered piezoelectric/electrostrictive film type device 20 shown in  
25 FIGS. 4 to 6, when the piezoelectric/electrostrictive materials are stacked among the electrodes 75 to 77 positioned in the lowermost and intermediate layers, or after

all the respective piezoelectric/electrostrictive layers 71, 72 are stacked, the piezoelectric/electrostrictive layers can be heat-treated at a predetermined temperature and obtained.

[0036] The piezoelectric/electrostrictive material for use

5 in the present invention may be crystalline or amorphous as long as field inducing strains such as piezoelectric or electrostrictive effects are caused after the heating treatment. Any of a semiconductor, ferroelectric ceramic, and antiferroelectric ceramic may be used in the

10 piezoelectric/electrostrictive material, and the material may appropriately be selected/used in accordance with the application.

[0037] Specific examples of the material include the ceramic containing one alone or two or more of lead zirconate, lead  
15 titanate, lead zirconate titanate, lead magnesium niobate, lead nickel niobate, lead zinc niobate, lead manganese niobate, lead antimony stannate, lead manganese tungstate, lead cobalt niobate, barium titanate, sodium bismuth titanate, potassium sodium niobate, and strontium bismuth tantalite.

20 [0038] Particularly, a material containing lead zirconate titanate (PZT-system) and/or lead magnesium niobate (PMN-system) as the major component, or sodium bismuth titanate as the major component is preferable, in that a stabilized composition having a high electromechanical coupling  
25 coefficient and piezoelectric constant and little reactivity with the ceramic substrate at the sintering time of the piezoelectric/electrostrictive film is obtained.

[0039] Furthermore, a material may also be used including the ceramic material containing a small amount of components to which at least one alone or two or more oxides of lanthanum, calcium, strontium, molybdenum, tungsten, barium, niobium, zinc, nickel, manganese, cerium, cadmium, chromium, cobalt, antimony, iron, yttrium, tantalum, lithium, bismuth, and tin are added. For example, by making lead zirconate, lead titanate, and/or lead magnesium niobate as the major components contain lanthanum or strontium, anti-electric field or piezoelectric property can sometimes be adjusted.

[0040] These piezoelectric/electrostrictive materials can be prepared, for example, by an oxide mixing method, and for example, material powder containing  $\text{PbO}$ ,  $\text{SrCO}_3$ ,  $\text{MgCO}_3$ ,  $\text{Nb}_2\text{O}_5$ ,  $\text{ZrO}_2$ , and the like can be weighed so as to obtain a predetermined composition, mixed, tentatively sintered, and crushed. The material can be prepared in this method. Examples of another method include a coprecipitation method, and alkoxide method.

[0041] Moreover, examples of the method of applying the piezoelectric/electrostrictive material include: various thick-film forming methods such as a screen printing method, dipping method, coat method, and electrophoretic migration method; and various thin-film forming methods such as an ion beam method, sputtering method, vacuum deposition method, ion plating method, chemical vapor deposition method (CVD), and plating. Above all, since the piezoelectric/electrostrictive layer 73 having satisfactory piezoelectric/electrostrictive



properties is obtained, the thick-film forming methods such as the screen printing method, dipping method, coat method, and electrophoretic migration method are preferable.

[0042] It is to be noted that the piezoelectric/

5 electrostrictive material may be printed or applied in an area broader than that of the applied portion of each electrodes in order to form the projecting portion 79. In the present invention, the projecting portion 79 may partially be bonded to the substrate 44. However, to avoid  
10 the reduction of the flexural displacement, preferably the projecting portion is not directly bonded to the substrate 44. To obtain this non-bonded state, for example, the substrate 44 is preferably formed of a material having little reactivity with the piezoelectric/electrostrictive material  
15 at the heating treatment time, such as zirconium oxide.

[0043] Moreover, when the multilayered piezoelectric/  
electrostrictive film type device 20 shown in FIGS. 4 to 6 is formed, for example, the electrode 77 is stacked on the substrate 44, and subsequently the piezoelectric/electro-  
20 strictive materials are stacked alternately with the plurality of electrodes 75, 76 by the above-described various methods.

[0044] In the present invention, the thickness of the layer formed of the piezoelectric/electrostrictive material is  
25 preferably set to be substantially equal to that of the substrate from the view point of securing the mechanical strength and desired flexural displacement of the device.

Specifically, a ratio of the thickness to that of the thin portion 66 of the substrate (the thin portion /a layer made of the piezoelectric/electrostrictive material) is preferably 0.1 to 30, more preferably 0.3 to 10, and particularly preferably 0.5 to 5.

[0045] When the ratio of the thickness to that of the substrate (the substrate/(the layer of piezoelectric/electrostrictive material)) is in this range, the substrate (the thin portion) easily follows the sintering shrinkage of the piezoelectric/ electrostrictive layer at a time of the subsequent heating treatment for forming the piezoelectric/electrostrictive layer after the coating of the substrate with the piezoelectric/electrostrictive material and; a dense piezoelectric/electrostrictive layer can be formed without causing the peel. A sufficient resistance to the vibration by the bending of the piezoelectric/electrostrictive layer can be imparted.

[0046] However, in order to realize the miniaturization of the device, the thickness of the layer of piezoelectric/ electrostrictive material is preferably 5 to 100  $\mu\text{m}$ , more preferably 5 to 50  $\mu\text{m}$ , and particularly preferably 5 to 30  $\mu\text{m}$ .

[0047] Moreover, in the case of the multilayered piezoelectric/electrostrictive film type device 20 shown in FIGS. 4 to 6, the piezoelectric/electrostrictive film type device is formed in a thin film shape, and an aspect ratio can be raised; and therefore, the thickness per layer formed of the piezoelectric/electrostrictive material is preferably

set to 30  $\mu\text{m}$  or less. Furthermore, a plurality of layers of piezoelectric/electrostrictive materials is preferably formed so that the layers gradually become thinner in order from the lower layer. For example, the layers are preferably formed

5 so that a thickness  $t_n$  of an n-th layer of piezoelectric/electrostrictive material from below satisfies the equation:  $t_n \leq t_{n-1} \times 0.95$ . Since the strain of the piezoelectric/electrostrictive layer is large at the same driving voltage when the piezoelectric/electrostrictive layer  
10 has a small thickness, the upper piezoelectric/electrostrictive layer is formed so that the layer is strained more than the lower piezoelectric/electrostrictive layer; thereby, a bending efficiency is improved, and the flexural displacement can more effectively be exhibited.

15 [0048] In the present invention, after forming the layer of the predetermined piezoelectric/electrostrictive material, the heating treatment of the layer of the piezoelectric/electrostrictive material is preferably carried out at a temperature of 1000 to 1400°C. In addition, the heating  
20 treatment is preferably carried out in the presence of an atmosphere controlling material having the same composition as that of the piezoelectric/electrostrictive material in order to obtain a ceramic composite which has a desired composition by avoiding volatilization of each component.

25 [0049] Next, in the manufacturing method of the present invention, as shown in FIG. 7, is applied a coating liquid 1 to 3 prepared by mixing a polymerizable oligomer and

inorganic particles in a dispersion medium, to make the coating liquid permeable through a gap between at least a projecting portion 79 of the piezoelectric/electrostrictive layer 73 and the substrate 44 in such a position and amount that does not coat at least a part of each electrode 75, 77; and then, this drawing shows a state immediately after the coating liquid 1 - 3 has been discharged, and just after this, the coating liquid permeates through the gap between the projecting portions 79 and the substrate 44, and then the resultant is subjected to drying step to form a coupling member to couple the projected portion of the piezoelectric/electrostrictive layer and the substrate.

[0050] Accordingly, the piezoelectric/electrostrictive film type device having a high resonance frequency can be obtained in a state in which the connection of the respective electrodes to the outside can easily be secured. In addition, an operation of peeling the coat layer formed on the electrode, which is necessary for the application by a spin coating method, is not required; and therefore, a secondary trouble caused by dust generated by the peeling can be prevented.

[0051] The aforementioned coating liquid for use in the present invention has a property of ease of flowing. In the application of the liquid, coating means capable of applying micro droplets of several to 1000 pL at a rate of several m/second to 20 m/second is preferably used. For example, as shown in FIGS. 8(a), 8(b), it is preferable to use a coating

apparatus including: a substrate 95 including a coating liquid introduction path 92 connected to a coating liquid supply source 81, a pressurizing chamber 93 in which the coating liquid introduction path 92 opens, and a plurality of coating liquid discharge paths 94 connected to the pressurizing chamber 93 and opened to the outside; and a piezoelectric/electrostrictive operation portion 96 disposed in a position corresponding to the pressurizing chamber 93 on the substrate 95; and an outer wall of the pressurizing chamber 93 is pushed in accordance with the flexural displacement of the piezoelectric/electrostrictive operation portion 96, and the coating liquid 1 introduced into the pressurizing chamber 93 is discharged in an atomized droplet state (hereinafter referred to as an "on-demand type coating apparatus"); or alternatively, as shown in FIG. 9, it is preferable to use a coating apparatus including: pressurizing supply means 101 for pressurizing/supplying the coating liquid 1; switching means 103 which is disposed in a supply path 102 of the pressurizing supply means 101 to switch the supply of the coating liquid 1; and a discharge head 104 for discharging the coating liquid 1 introduced from the supply path 102 of the pressurizing supply means 101 to the outside; and the discharge head 104 includes: a substrate 108 including a coating liquid introduction path 105 connected to the supply path 102 of the pressurizing supply means 101, a pressurizing chamber 106 in which the coating liquid introduction path 105 opens, and a plurality of coating

liquid discharge paths 107 connected to the pressurizing chamber 106 and opened to the outside; and a piezoelectric/electrostrictive operation portion 109 disposed in a position corresponding to the pressurizing chamber 106 on the substrate 108. When the switching means 103 is opened, the outer wall of the pressurizing chamber 93 is pushed by the flexural displacement of the piezoelectric/electrostrictive operation portion 109, and the coating liquid introduced into the pressurizing chamber 106 is continuously discharged in the atomized droplet state in the coating apparatus (hereinafter referred to as "the continuous coating apparatus").

[0052] According to the on-demand type coating apparatus, since the coat amount can precisely be controlled, and a coating timing is precise, a predetermined amount of solution can precisely be supplied to a predetermined position, coat position accuracy is improved, and a coat film thickness can further be uniformed.

[0053] First, according to the continuous coating apparatus, it is easy to apply a large amount of a highly viscous solution every small amount by the pressurizing supply means. Therefore, the coating liquid little spreads over the substrate, a coat pattern is miniaturized, and a small amount of droplets can be supplied in a large amount, a coating time can therefore be reduced, and the device can be produced with good efficiency. Since the solution is pressurized, a probability that coating is impossible because of a dried

coating liquid in a nozzle can be reduced, and stable, coating is possible. It is to be noted that in FIG. 8(b) and 9(b), arrows show channels of the coating liquid.

[0054] Furthermore, in the continuous coating apparatus, a regulator 121 for pressure adjustment is also preferably disposed between the pressurizing supply means 101 and switching means 103 from the viewpoint of improving stability of the coating liquid amount. For the enhancement of response of the coating liquid, pressure release means 122 is similarly preferably disposed between the pressurizing supply means 101 and switching means 103.

[0055] Additionally, in the present invention, when the continuous coating apparatus or the on-demand type coating apparatus is used, it is preferable to fix the piezoelectric/electrostrictive film type device 10 on an XY stage 4 shown in FIG. 9(a) and apply the coating liquid from the viewpoint of easy application of the coating liquid to the predetermined position.

[0056] Moreover, in the present invention, the on-demand type fine coating apparatus is preferably used in combination with the continuous fine coating apparatus so that the liquid can be applied in a very fine pattern and can efficiently be applied in a broader range. For example, when the coating liquid 1 - 3 is applied, as shown in FIG. 7, the coating liquid 2 for the application in the broad range is applied preferably with a continuous coating apparatus and the coating liquid 1 and 3 for the formation of a finer pattern

is applied preferably with an on-demand type coating apparatus.

[0057] More specifically, in a peripheral edge portion of the piezoelectric/electrostrictive operation portion 78 in which the gap between the piezoelectric/electrostrictive layer 73 and substrate 44 is open, on the portion being located at a substantially equal distance from the terminal portion 75a of the upper electrode 75 and the terminal portion 77a of the lower electrode is applied a large amount of a coating liquid 2 with the continuous coating apparatus; and in the peripheral edge of the piezoelectric/electrostrictive operation portion 78, on the positions being located at the vicinity of the terminal portion 75a of the upper or lower electrode (within 0.5 mm from the terminal portion 75a) is preferably applied a small amount of the coating liquid 1, 3 with an on-demand type coating apparatus.

[0058] When the coating methods are combined in this manner, a large amount of coating liquid is diffused in the periphery and can efficiently be applied from a position most distant from the terminal portion 75a of the upper electrode 75 and the terminal portion 77a of the lower electrode 77; and moreover, the small amount of coating liquid is applied in the vicinity of the terminal portions 75a, 77a of the upper electrode 75 and lower electrode 77; thereby in the gap between the projecting portion of the piezoelectric/electrostrictive layer and the substrate, the coating liquid can also permeate through the portions in the vicinity of the



terminal portions 75a, 77a of the upper electrode 75 and lower electrode 77 in the same manner as in the other portions while avoiding the coating of the respective electrodes 75, 77. Thus, while the conduction of each electrode is secured, the piezoelectric/electrostrictive film type device having the desired coupling member material formed in the gap between the projecting portion of the piezoelectric/electrostrictive layer and the substrate and having a high resonance frequency can be manufactured.

[0059] Moreover, in the present invention, as shown in FIG. 10, positions and sizes of opening of coating liquid discharge paths 94a, 94b, 94c in the coating apparatus are preferably adjusted so that a desired amount of coating liquid is applied on a desired portion (FIG. 10 shows the on-demand type coating apparatus, but the continuous coating apparatus is also basically the same). In this case, the nozzle positions and sizes of the respective coating liquid discharge paths 94a, 94b, 94c do not have to be necessarily adjusted so as to open the respective coating liquid discharge paths 94a, 94b, 94c at equal intervals and to obtain the same aperture among the respective coating liquid discharge paths, and it is also preferable to set different apertures or positions of the respective coating liquid discharge paths 94a, 94b, 94c among discharge heads or in the same discharge head.

[0060] Accordingly, it is possible to simultaneously coat a portion that requires pattern precision and a portion that

does not require the precision, and the patterning of desired coatings can efficiently be made. More specifically, in consideration of mutual influence of the aperture and position, the nozzle position or size may be adjusted so that the coating liquid can permeate through the gap between the projecting portion of the piezoelectric/electrostrictive layer and the substrate, and complete coating of each electrode can be avoided. For example, as shown in FIG. 7, in the peripheral edge of the piezoelectric/electrostrictive operation portion 78 in which there is the gap between the piezoelectric/electrostrictive layer 73 and substrate 44, the coating liquid discharge path having a largest aperture is disposed in the portion positioned substantially at the equal distance from the terminal portion 75a of the upper electrode 75 and from the terminal portion 77a of the lower electrode 77, and the portion is coated with a largest amount of coating liquid 2; and in the peripheral edge of the piezoelectric/electrostrictive operation portion 78, the portions positioned in the vicinity of the terminal portion 77a of the upper or lower electrode (within 0.5 mm from each terminal portion 75a) are preferably coated with a small amount of coating liquid 1, 3 by opening the coating liquid discharge paths having a smallest aperture.

[0061] When the amount and position to be coated in this manner, the large amount of coating liquid is diffused and can efficiently be applied from the position most distant from the terminal portion 75a of the upper electrode 75 and

from the terminal portion 77a of the lower electrode 77, and  
by the application of the small amount in the vicinity of the  
terminal portions 75a, 77a of the upper electrode 75 and  
lower electrode 77, in the gap between the projecting portion  
5 of the piezoelectric/electrostrictive layer and the substrate,  
the coating liquid can permeate through the portions in the  
vicinity of the terminal portion 75a of the upper electrode  
75 and the terminal portion 77a of the lower electrode 77 in  
the same manner as in the other portion while avoiding the  
10 coating of the terminal portions 75a of the respective  
electrodes. Therefore, while the conduction of each  
electrode is secured, the piezoelectric/electrostrictive film  
type device having the desired coupling member in the gap  
between the projecting portion of the piezoelectric/  
15 electrostrictive layer and the substrate and having the high  
resonance frequency can be manufactured.

[0062] Moreover, in the present invention, in addition to  
the adjustment of the coat position and amount, a discharge  
apparatus having a plurality of coating liquid discharge  
20 paths, a discharge apparatus having a plurality of discharge  
heads, or a discharge apparatus having a plurality of  
discharge heads having a plurality of coating liquid  
discharge paths may be used to change the viscosity or  
composition of the coating liquid introduced into each  
25 coating liquid discharge path by each coating liquid  
discharge path for application of the coating liquid having a  
different viscosity or composition by each coat position.

For example, as shown in FIG. 11, it is preferred that, in the peripheral edge of the piezoelectric/electrostrictive operation portion 78 including the gap opening between the piezoelectric/electrostrictive layer 73 and substrate 44, a coating liquid 5 having a lowest viscosity is discharged to the portion positioned substantially at the equal distance from the terminal portion 75a of the upper electrode 75 and the terminal portion 77a of the lower electrode 77; and in the peripheral edge of the piezoelectric/electrostrictive operation portion 78, coating liquids 4, 6 having a high viscosity are discharged to coat the portion positioned in the vicinity of the terminal portions 75a, 77a of the upper electrode 75 and lower electrode 77 (within 0.5 mm from each terminal portion 75a). By thus changing the viscosity of the coating liquid in accordance with the coat position to apply the coating liquid having the low viscosity to the position most distant from the terminal portion 75a of the upper electrode 75 and the terminal portion 77a of the lower electrode 77 and so as to enable further diffusing in the periphery; and to apply the coating liquid having the high viscosity to the position in the vicinity of the terminal portion 75a of the upper electrode 75 and the terminal portion 77a of the lower electrode 77, while the diffusion of the coating liquid having the low viscosity is inhibited, the coating liquid is allowed to permeate through even the portions in the vicinity of the terminal portions 75a, 77a of the upper electrode 75 and lower electrode 77 in the gap

between the projecting portion of the piezoelectric/electrostrictive layer and the substrate in the same manner as in the other portions, and moreover, the coating of each electrode 75, 77 can be avoided.

5 [0063] Additionally, in the present invention, when a coupling member 70 for connecting the projecting portion 79 of the piezoelectric/electrostrictive layer 73 to the substrate 44 is disposed without any large drying shrinkage, the viscosity of the coating liquid is preferably 1000 cP or  
10 less, more preferably 300 cP or less, particularly preferably 50 cP or less. When the on-demand type coating apparatus and continuous coating apparatus are used, a viscosity of 20 cP or less is particularly preferable.

[0064] Moreover, in the manufacturing method of the present  
15 invention, as shown in FIGS. 14(a) and 14(b), when a piezoelectric/electrostrictive film type device is manufactured in which pores opening in the piezoelectric/electrostrictive layer 73 (including a contact surface of the piezoelectric/electrostrictive layer 73 with  
20 the upper electrode 75) or the upper electrode 75 are filled with the coating liquid or when a piezoelectric/electrostrictive film type device is manufactured in which a micro restraint layer 90 of the same material as that of the coupling member 70 is disposed in a predetermined thickness  
25 to coat the contact surface of the piezoelectric/electrostrictive layer 73 with the upper electrode 75 or the upper electrode 75, the surface of the applied portion of the upper

electrode 75 may also be coated with the coating liquid.

[0065] However, since a binding force needs to be minimized, the thickness is preferably  $1/15$  or less with respect to a total thickness of the thin portion 66 and

5 piezoelectric/electrostrictive operation portion, more preferably  $1/30$  or less. Both the on-demand type coating apparatus and the continuous coating apparatus can preferably be used. When a thinner film is prepared in the present apparatus, the viscosity is set to  $0.5$  to  $5$  cP, and a  
10 dropping rate is preferably set to  $3$  m/second or more.

Furthermore, when only an opened pores portion is filled with the coating liquid, compressed air may be sprayed after the coating. Incidentally, in the piezoelectric/electrostrictive film type device provided with the filled micro-pores or the  
15 micro restraint layer 90, insulation of the piezoelectric/electrostrictive operation portion can be enhanced.

[0066] In the present invention, the coating liquid is preferably applied while vibrating at least the

20 piezoelectric/electrostrictive layer or the substrate. Since the coating liquid can securely be supplied between the substrate and piezoelectric/electrostrictive layer without including any bubble, the substrate can more firmly be connected to the piezoelectric/electrostrictive layer.

25 [0067] In this case, examples of vibrating means include: vibrating means including an ultrasonic vibrator 110 disposed on the stage 4 to which the piezoelectric/electrostrictive

film type device 10 is fixed to mainly vibrate the substrate 44 as shown in FIG. 9(a); or vibrating means for applying a voltage to the piezoelectric/electrostrictive operation portion of the piezoelectric/electrostrictive film type device 10 from an external power source (not shown) to vibrate the piezoelectric/electrostrictive operation portion.

[0068] As the coating liquid for use in forming the coupling member in the present invention, a coating liquid is preferable in which inorganic particles and polymerizable oligomer are mixed in dispersion liquid. When this coating liquid is used, the coupling member can be made of a hybrid material including the inorganic particles scattered in a matrix of a polymer compound, and it is possible to obtain the piezoelectric/electrostrictive film type device which has the flexural displacement equal to that of the conventional piezoelectric/electrostrictive film type device and which is superior in high-speed response. In a drying step performed in forming the coupling member, shrinkage of the coupling member is suppressed, and the coupling member or the piezoelectric/electrostrictive layer can be prevented from being cracked. Incidentally, in the present invention, it is possible to use the coating liquid containing the above-described polymer compound mixed in the dispersion liquid from the beginning, but the coating liquid having the polymerizable oligomer mixed in the dispersion liquid is preferable in order to form the coupling member having preferable properties.

[0069] In the present invention, examples of a dispersion medium for use in the coating liquid include water, methanol, ethanol, propanol, isopropyl alcohol, butanol, and acetone from the viewpoint that a dispersion liquid having a homogeneous polar dispersion medium is easily obtained.

[0070] Moreover, examples of the inorganic particles mixed in the dispersion medium preferably include particles of an oxide containing at least one element selected from Ti, Zr, V, Nb, Cr, Mo, W, Al, Mn, Fe, Co, Ni and Si. These inorganic particles can be used as one type alone or as a combination of two or more types.

[0071] Moreover, an average particle diameter of the inorganic particles is preferably 5 nm to 1  $\mu$ m, more preferably 10 nm to 200 nm. When the average particle diameter is less than 5 nm, an effect of suppressing the shrinkage of the coupling member is small in the drying step performed in forming the coupling member; and on the other hand, when the average particle diameter exceeds 1  $\mu$ m, the inorganic particles are easily precipitated in the coating liquid, and it is difficult to obtain the homogeneous coupling member.

[0072] Moreover, if the average particle diameter of the inorganic particles is in this range, it is possible to satisfy a requirement that restraint of the coupling member with respect to the piezoelectric/electrostrictive layer is avoided and a requirement that rigidity of the piezoelectric/electrostrictive layer 73 is increased to enhance the high-



speed response with good balance.

[0073] Furthermore, the inorganic particles further have a two-peaks type particle size distribution. A ratio (D/C) of an average particle diameter (C) of large-diameter inorganic particles having a diameter larger than that corresponding to an inflection point existing between two peaks to an average particle diameter (D) of small-diameter inorganic particles having a diameter smaller than that corresponding to the inflection point is preferably 0.05 to 0.7, more preferably 0.1 to 0.5.

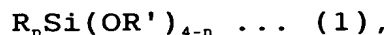
[0074] When the inorganic particles have the two-peaks type particle size distribution, voids among the large-diameter inorganic particles can be filled with the small-diameter inorganic particles to raise a volume fraction of the inorganic particles in the coupling member; and therefore, in the drying step accompanying in the formation of the coupling member, the shrinkage of the coupling member can further be reduced, the rigidity of the piezoelectric/electrostrictive layer 73 can further be increased, and the high-speed response can further be enhanced.

[0075] Moreover, for the inorganic particles that have the two-peaks type particle size distribution, a ratio (F/E) of a mass (E) of the large-diameter inorganic particles to a mass (F) of the small-diameter inorganic particles is preferably 0.05 to 0.7, more preferably 0.1 to 0.5 with similar respect.

[0076] Furthermore, examples of the polymerizable oligomer in the present invention include a polymerizable oligomer for

a vinyl polymer such as an acryl resin, addition polymer such as an epoxy resin and polyurethane, condensed polymer such as polyester and polycarbonate, and polysiloxane polymer (polymer having repeating unit of Si-O-Si) which is polymer of an organic silicon compound; and above all, from resistances to heat, water, and chemical, and water repellency, the polymerizable oligomer in which several to several tens of monomers shown in the following general formula (1) are condensation-polymerized is preferable.

[0077]



where R and R' denote the same type or different types of organic groups, and n is an integer of 0 to 3.

[0078] In the present invention, in the above general

formula (1), R includes alkyl groups such as a methyl group, ethyl group, and propyl group, aryl groups such as a phenyl group, alkenyl groups such as a vinyl group, and substituted alkyl groups such as a  $\gamma$ -methacryloxypropyl group,  $\gamma$ -glycidoxypropyl group,  $\gamma$ -chloropropyl group,  $\gamma$ -mercaptopropyl group,  $\gamma$ -aminopropyl group, and trifluoromethyl group. In addition, include the alkyl groups such as the methyl group, ethyl group, propyl group, and butyl group, the aryl groups such as the phenyl group, and the substituted alkyl groups such as a  $\beta$ -methoxy ethoxy group, and acetyl group.

[0079] Incidentally, in the present invention, the polymerizable oligomer is condensation-polymerized, for example, by dehydration or dealcoholization reaction by the

drying described later, and accordingly desired polymer compounds constituting the matrix of the coupling member may be obtained.

[0080] In the present invention, a blend ratio of the polymerizable oligomer to the inorganic particles is preferably set to be in an appropriate range in accordance with the type of each component. For example, with the use of the polymerizable oligomer obtained by condensation-polymerizing the monomers shown in the general formula (1), it is preferable to contain the inorganic particles in the coating liquid at a ratio of 0.1 to 300 parts by weight with respect to 100 parts by weight of the polymerizable oligomer; and it is more preferable to contain the inorganic particles in the coating liquid at a ratio of 1 to 100 parts by weight.

[0081] In the present invention, for the coating liquid, it is possible to combine the polymerizable oligomer corresponding to various types of polymer compounds described above with various types of inorganic particles. Above all, the coating liquid preferably contains the polymerizable oligomer corresponding to polysiloxane polymer and the silica particles, more preferably contains the polymerizable oligomer shown in the above general formula (I) and silica particles.

[0082] When the coupling member 70 is formed with the coating liquid of the composition, there can be obtained a piezoelectric/electrostrictive film type device having high rigidity of the piezoelectric/electrostrictive layer 73

without reducing the flexural displacement of the thin portion 66 and piezoelectric/electrostrictive layer 73; and furthermore, since the tenacity of the formed coupling member 70 is high, a resistance to high-speed repeated driving of the piezoelectric/electrostrictive film type device may be enlarged.

[0083] In the present invention, after applying the coating liquid, the coupling member is formed by drying. In the drying, appropriate conditions are preferably selected in accordance with the type of composition of the coating liquid. For example, in the case that the coating liquid is mixed with the polymerizable oligomer in accordance with the vinyl polymers such as the acryl resin, the addition polymers such as the epoxy resin and polyurethane, or the condensation polymers such as polyester and polycarbonate, the layer is left to stand at room temperature after applying the coating liquid, and the drying may be performed.

[0084] On the other hand, in the case that the polymerizable oligomer corresponding to the aforementioned polysiloxane polymer is mixed in the coating liquid; it is preferred that, after applying the aforementioned coating liquid, the layer is left to stand for ten or more minutes at the room temperature, to move a large part of solvent, and thereafter an atmospheric temperature is raised at a desired temperature at a rate of 600°C/Hr or less preferably to perform the drying by heating.

[0085] When the heating and drying are performed

immediately after the coating, or the temperature is rapidly raised to perform the drying, because of rapid evaporation of the solvent in the coating liquid, the coating liquid rapidly contracts, which sometimes cause cracks in the coupling member, or exfoliation at an interface between the coupling member and piezoelectric/electrostrictive layer.

[0086] Moreover, the drying by the heating is performed preferably at 60 to 120°C, more preferably at 100 to 120°C. The drying at a temperature exceeding 120°C, for the similar reasons as those of the aforementioned leaving to stand at room temperature, the coating liquid rapidly contracts because of rapid evaporation of the solvent in the coating liquid, which sometimes cause cracks in the coupling member, or exfoliation at the interface between the material and the piezoelectric/electrostrictive layer. On the other hand, when the drying is performed at a temperature lower than the heating temperature, the organic solvent and moisture dissolved in the solvent are insufficiently removed.

[0087] When the coating liquid mixed with the polymerizable oligomer corresponding to the polysiloxane polymer is applied, further after the drying, a hardening treatment by the heating at a higher temperature is preferably performed continuously or in a separate step. To be more specific, the heating preferably at a temperature of 700°C or less is preferable, the heating at a temperature of 600°C or less is more preferable, the heating at a temperature less than 500°C is further preferable, and the heating at a temperature less

than 450°C is particularly preferable.

[0088] When the temperature of the hardening treatment by the heating exceeds the above-described temperature range, the components such as Si in the coupling member reacts with the material constituting the piezoelectric/electrostrictive layer to deteriorate capabilities of the piezoelectric/electrostrictive layer, and since defects are generated in the piezoelectric/electrostrictive layer in some cases, dielectric or mechanical breakdown sometimes occurs. In addition, the organic components in the coupling member are decomposed to cause a crack in the coupling member.

[0089] Incidentally, in the present invention, by adjusting a constituting ratio of the polymerizable oligomer to the inorganic particles in the coating liquid, or by adjusting the heating temperature at the hardening treatment time to change a bond strength of Si-O-Si bond, it is possible to optimize mechanical properties such as hardness (hardness rises as the temperature rises), and chemical properties such as water repellency.

[0090] The piezoelectric/electrostrictive film type device and the manufacturing method of the device according to the present invention have been described above, but the piezoelectric/electrostrictive film type device obtained by the manufacturing method of the present invention can be used, for example, as a driving portion 120 of a display device or ink jet printer head as shown in FIGS. 12, 13. Specifically, it is possible that, as shown in FIG. 12, with a light wave-

guide plate 200 for introducing a light 180 from a light source 160, the driving portion 120 having the piezoelectric/electrostrictive film type device 10 of the present invention as a main constituting element is disposed so as to face the rear surface of the light wave-guide plate 200, and in a position corresponding to a pixel; and furthermore, pixel constituting materials 30 are stacked on the driving portion 120, and the pixel constituting materials 130 is brought into contact with or separated from the light wave-guide plate 200 by a driving operation of the driving portion 120 to constitute the display device. Moreover, it is possible that, as shown in FIG. 13, the driving portion 120 including the piezoelectric/electrostrictive film type device 10 of the present invention as the main constituting element and including a pressurizing chamber 100 constituted of the cavity 48 of the substrate 44 is integrally bonded to an ink nozzle member 111 including a nozzle hole 112 opened to the outside from the pressurizing chamber 100 of the driving portion 120 through a channel for ink jet 117, and an orifice hole 114 opened into the pressurizing chamber 100 of the driving portion 120 from an ink supply source through a channel for ink supply 118, to constitute the ink jet printer head. Incidentally, as to the specific construction of the display device or the ink jet printer head, JP-A-2001-343598 and JP-A-11-147318 will be cited here.

[0091]

(Example) The present invention will more specifically be

described hereinafter by examples of a piezoelectric film type device, but the present invention is not limited to these examples. Incidentally, evaluation was performed as follows with respect to each example and comparative example.

5 [0092]

(Evaluation Method)

(1) Flexural Displacement

The displacement at the time of applying an electric field of 3 kV/mm was applied to each piezoelectric film type device obtained in examples and comparative examples at room temperature was measured using a laser Doppler vibration meter.

[0093]

(2) Resonance Frequency

15 A resonance frequency of the piezoelectric film type device obtained in each example or comparative example was measured with the laser Doppler vibration meter and FFT analyzer.

Specifically, a swept sine waveform generated by the FFT analyzer (waveform containing a plurality of frequency components) was applied to the device to drive the device. The vibration of the device was measured with the laser Doppler vibration meter, a speed output of the laser Doppler vibration meter was inputted into the FFT analyzer to analyze the frequency, and a lowest peak was regarded as the resonance frequency.

25 [0094]



(Example 1)

A platinum-made lower electrode (dimension: 1.7×0.8 mm (in the length of 1.7 mm, 0.3 mm corresponds to the terminal portion), thickness: 3 μm) was formed on a substrate whose thin portion and fixing portion were both made of Y<sub>2</sub>O<sub>3</sub>-stabilized ZrO<sub>2</sub> (dimension of the thin portion: 1.6×1.1 mm, thickness: 10 μm) by the screen printing method. The heat treatment at a temperature of 1300°C for 2 hours was carried out to unitarily join the electrode with the substrate.

[0095] Thereon, a piezoelectric material constituted of (Pb<sub>0.999</sub>La<sub>0.001</sub>)(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)<sub>0.375</sub>Ti<sub>0.375</sub>Zr<sub>0.250</sub>O<sub>3</sub> in which a part of Pb was substituted with 0.1 mol% of La (average particle diameter of 0.49 μm, maximum particle diameter of 1.8 μm) was laminated in a thickness of 20 μm in a broader range of 1.3×0.9 mm including the surface corresponding to the upper surface of the lower electrode by the screen printing method.

[0096] Subsequently, the atmosphere-controlling material having the same composition as that of the piezoelectric material was placed in a container together with the piezoelectric material, and the laminate of the piezoelectric material on the electrode-formed substrate was heat-treated at 1275°C for two hours. The thickness of the piezoelectric layer after the heat treatment was 13 μm.

[0097] Subsequently, the upper electrode of gold was formed in a thickness of 0.5 μm in a range of 1.7mm×0.8 mm (in the length of 1.7 mm, 0.3 mm corresponds to the terminal portion) on the piezoelectric layer by the screen printing method, and

heat-treated at 600°C.

[0098] Subsequently, thus obtained device was fixed onto a sample stage after covering its surface opposite to the one in which the piezoelectric/electrostrictive layer and

5 electrode had been disposed with a UV sheet, and 30% by mass of colloidal silica having two-peaks particle size distribution and containing in a mixed state a large-diameter amorphous silica having an average particle diameter of 100 nm larger than an inflection point of 50 nm existing between  
10 two peaks and a small-diameter amorphous silica having an average particle diameter of 20 nm smaller than the inflection point of 50 nm were mixed with 70% by mass of a polymerizable oligomer solution having been prepared by mixing 20% by mass of an equimolar mixture of

15 tetraethoxysilane and methylethoxysilane with a mixed dispersion medium of isopropyl alcohol and water to prepare a coating liquid, which was applied using the on-demand type coating apparatus provided with three coating liquid discharge paths. In this case, the apertures of the  
20 respective coating liquid discharge paths were set to  $\phi 0.03$  mm,  $\phi 0.07$  mm, and  $\phi 0.03$  mm. Regarding a position of opening of each coating liquid discharge path, the coating liquid discharge path having an aperture of  $\phi 0.07$  mm was set to a position substantially at an equal distance from the terminal  
25 portions of the upper electrode 75 and lower electrode 77 in the piezoelectric/electrostrictive operation portion peripheral edge (on both sides of the thin portion in the

direction of width) in which the gap between the piezoelectric/electrostrictive layer and substrate is opened, and the coating liquid discharge path having an aperture of  $\phi 0.03$  mm was set to a position of 0.45 mm on the both sides of the thin portion in a longitudinal direction. The discharge amount per droplet was set to 200 pL/droplet in the coating liquid discharge path having an aperture of  $\phi 0.07$  mm, and 70 pL/droplet in the coating liquid discharge path having an aperture of  $\phi 0.03$  mm. Colloidal silica in which large-diameter amorphous silica was mixed with small-diameter amorphous silica was prepared by mixing and stirring 70% by mass of colloidal silica (solid concentration of 20% by mass) in which large-diameter amorphous silica having an average particle diameter of 100 nm was dispersed in isopropyl alcohol with 30% by mass of colloidal silica (solid concentration of 20% by mass) in which small-diameter amorphous silica having an average particle diameter of 20 nm was dispersed in isopropyl alcohol.

[0099] Finally, the piezoelectric/electrostrictive film type device coated with the coating liquid was left to stand at room temperature for 30 minutes. Thereafter, the temperature was raised at a temperature rise rate of 200°C/h, held at 80°C to 120°C for one hour, and continuously raised at 300°C. At the temperature, the hardening treatment was performed for 60 minutes, the coupling member for connecting the whole projecting portion of the piezoelectric/electrostrictive layer to the substrate was hardened, and the

piezoelectric/electrostrictive film type device was manufactured.

[0100] The obtained piezoelectric/electrostrictive film type device had a flexural displacement of 0.14  $\mu\text{m}$ , and a resonance frequency of 1.65 MHz. The surface of a conductive portion of each electrode of the piezoelectric/electrostrictive film type device remained exposed, and was ready for solder bonding of conductive wires to the electrode.

[0101]

10 (Comparative Example)

The piezoelectric/electrostrictive film type device was manufactured in the same manner as in Example 1 except that the whole piezoelectric/electrostrictive film type device was coated with the coating liquid using a spin coating apparatus instead of the discharge apparatus.

[0102] The obtained piezoelectric/electrostrictive film type device had a flexural displacement of 0.14  $\mu\text{m}$ , and a resonance frequency of 1.65 MHz. However, in the obtained piezoelectric/electrostrictive film type device, the conductive portion of each electrode was coated and was not ready for the solder bonding, and therefore, it was necessary to remove the coat layer in order to secure the conduction.

[0103] As described above, according to the present invention, there can be provided a manufacturing method of a piezoelectric/electrostrictive film type device, in which the piezoelectric/electrostrictive film type device capable of obtaining a high resonance frequency and superior in a high-

speed response can efficiently be manufactured while securing conduction of electrodes.

WHAT IS CLAIMED IS:

1. A manufacturing method of a piezoelectric/  
electrostrictive film type device including a  
piezoelectric/electrostrictive operation portion containing a  
5 lower electrode, a piezoelectric/ electrostrictive layer, and  
an upper electrode being stacked on a substrate of ceramic,  
wherein said method comprises the steps of:

forming the piezoelectric/electrostrictive layer of  
the piezoelectric/electrostrictive operation portion in a  
10 range broader than that of an applied portion of each  
electrode to project an end of the piezoelectric/electro-  
strictive layer;

coating a coating liquid prepared by admixing a  
polymerizable oligomer and inorganic particles in a  
15 dispersion medium in an amount sufficient to make the coating  
liquid permeable through a gap between at least a projecting  
portion of the piezoelectric/electrostrictive layer and the  
substrate, and coat an each electrodes in such a position and  
an amount that does not coat at least a part of each  
20 electrode; and

drying the coating liquid to form a coupling member  
to couple the projected portion of the  
piezoelectric/electrostrictive layer to the substrate.

25 2. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 1, using  
a coating apparatus comprising: pressurizing supply means for

pressurizing/supplying the coating liquid; switching means which is disposed in a supply path of the pressurizing supply means to switch the supply of the coating liquid; and a discharge head for discharging the coating liquid introduced from the supply path of the pressurizing supply means to the outside, the discharge head comprising: a substrate including a coating liquid introduction path communicating with the supply path of the pressurizing supply means, a pressurizing chamber in which the coating liquid introduction path opens, and one or more coating liquid discharge paths communicating with the pressurizing chamber and opened to the outside; and a piezoelectric/electrostrictive operation portion disposed in a position corresponding to the pressurizing chamber on the substrate, wherein at an open time of the switching means, the coating liquid introduced into the pressurizing chamber is continuously discharged in an atomized droplet state by flexural displacement of the piezoelectric/electrostrictive operation portion.

3. The manufacturing method of the piezoelectric/electrostrictive film type device according to claim 1, using a coating apparatus comprising: a substrate including a coating liquid introduction path communicating with a coating liquid supply source, a pressurizing chamber in which the coating liquid introduction path is opened, and one or more coating liquid discharge paths communicating with the pressurizing chamber and opened to the outside; and a

piezoelectric/electrostrictive operation portion disposed in  
a position corresponding to the pressurizing chamber, wherein  
in accordance with flexural displacement of the  
piezoelectric/electrostrictive operation portion, the coating  
5 liquid introduced into the pressurizing chamber is discharged  
in an atomized droplet state.

4. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 2,  
10 applying the coating liquid in an amount which differs  
depending on a position to be applied using any one of a  
coating apparatus comprising a discharge head including a  
plurality of coating liquid discharge paths having different  
apertures, and a coating apparatus comprising a plurality of  
15 discharge heads different from one another in the aperture of  
the coating liquid discharge path.

5. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 3,  
20 applying the coating liquid in an amount which differs  
depending on a position to be applied using a coating  
apparatus comprising a plurality of coating liquid discharge  
paths having different apertures.

25 6. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 1,  
applying the coating liquid with vibrating at least the



substrate or the piezoelectric/electrostrictive layer.

7. The manufacturing method of a piezoelectric/  
electrostrictive film type device provided with a  
5 piezoelectric/electrostrictive operation portion in which a  
plurality of electrodes and a plurality of  
piezoelectric/electrostrictive layers are stacked on a  
substrate of ceramic, wherein said method comprises the steps  
of:

10 forming each piezoelectric/electrostrictive layer of  
the piezoelectric/electrostrictive operation portion in a  
range broader than that of an applied portion of each  
electrode to project an end of the piezoelectric/electro-  
strictive layer;

15 coating a coating liquid prepared by admixing a  
polymerizable oligomer and inorganic particles in a  
dispersion medium in an amount sufficient to make the coating  
liquid permeable through a gap between at least a projecting  
portion of the piezoelectric/electrostrictive layer and the  
20 substrate, and coat an each electrodes in such a position and  
an amount that does not coat at least a part of the  
electrodes; and

drying the coating liquid to form a coupling member  
to couple the projected portion of the piezoelectric/electro-  
25 strictive layer to the substrate.

8. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 7, using  
a coating apparatus comprising: pressurizing supply means for  
pressurizing/supplying the coating liquid; switching means  
5 which is disposed in a supply path of the pressurizing supply  
means to switch the supply of the coating liquid; and a  
discharge head for discharging the coating liquid introduced  
from the supply path of the pressurizing supply means to the  
outside, the discharge head comprising: a substrate including  
10 a coating liquid introduction path communicating with the  
supply path of the pressurizing supply means, a pressurizing  
chamber in which the coating liquid introduction path opens,  
and one or more coating liquid discharge paths communicating  
with the pressurizing chamber and opened to the outside; and  
15 a piezoelectric/electrostrictive operation portion disposed  
in a position corresponding to the pressurizing chamber on  
the substrate, wherein at an open time of the switching means,  
the coating liquid introduced into the pressurizing chamber  
is continuously discharged in an atomized droplet state by  
20 flexural displacement of the piezoelectric/electrostrictive  
operation portion.

9. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 7, using  
25 a coating apparatus comprising: a substrate including a  
coating liquid introduction path communicating with a coating  
liquid supply source, a pressurizing chamber in which the

coating liquid introduction path is opened, and one or more coating liquid discharge paths communicating with the pressurizing chamber and opened to the outside; and a piezoelectric/electrostrictive operation portion disposed in a position corresponding to the pressurizing chamber, wherein in accordance with flexural displacement of the piezoelectric/electrostrictive operation portion, the coating liquid introduced into the pressurizing chamber is discharged in an atomized droplet state.

10

10. The manufacturing method of the piezoelectric/electrostrictive film type device according to claim 8, applying the coating liquid in an amount which differs depending on a position to be applied using any one of the coating apparatus comprising a discharge head including a plurality of coating liquid discharge paths having different apertures, and a coating apparatus comprising a plurality of discharge heads different from one another in the aperture of the coating liquid discharge path.

20

11. The manufacturing method of the piezoelectric/electrostrictive film type device according to claim 9, applying the coating liquid in an amount which differs depending on a position to be applied using a coating apparatus comprising a plurality of coating liquid discharge paths having different apertures.

25

12. The manufacturing method of the piezoelectric/  
electrostrictive film type device according to claim 7,  
applying the coating liquid with vibrating at least the  
substrate or the piezoelectric/electrostrictive layer.

# ABSTRACT OF THE DISCLOSURE

There is provided a manufacturing method of a piezoelectric/electrostrictive film type device, capable of effectively manufacturing a piezoelectric/electrostrictive film type device which has flexural displacement and durability equal to or more than those of a conventional piezoelectric/electrostrictive film type device and is superior in high-speed response while conduction of electrodes is secured. In the manufacturing method of a piezoelectric/electrostrictive film type device includes a piezoelectric/electrostrictive operation portion containing a lower electrode, a piezoelectric/ electrostrictive layer, and an upper electrode being stacked on a substrate of a ceramic, the method comprises the steps of, forming the piezoelectric/electrostrictive layer of the piezoelectric/electrostrictive operation portion in a range broader than that of an applied portion of each electrode to project an end of the piezoelectric/electrostrictive layer, coating a coating liquid prepared by admixing a polymerizable oligomer and inorganic particles in a dispersion medium in an amount sufficient to make the coating liquid permeable through a gap between at least a projecting portion of the piezoelectric/electrostrictive layer and the substrate, and coat an each electrodes in such a position and an amount that does not coat at least a part of each electrode, and drying the coating liquid to form a coupling member to couple the projected portion of the piezoelectric/electrostrictive layer

to the substrate.

FIG. 1

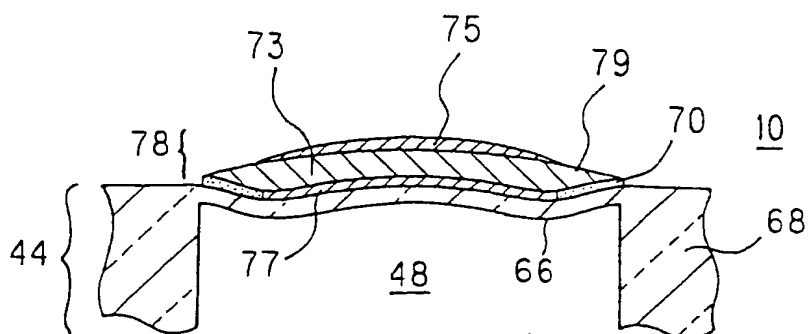


FIG. 2

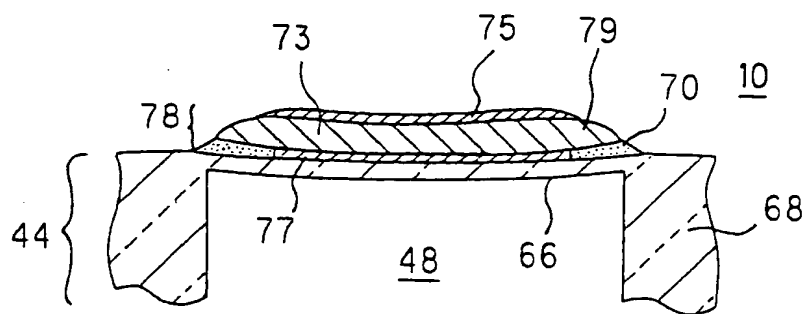


FIG. 3

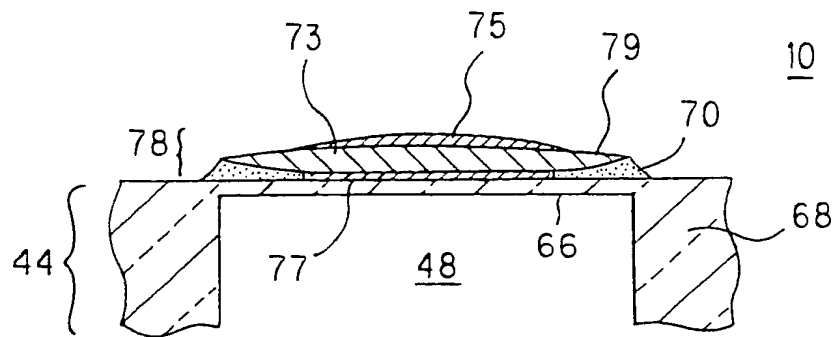


FIG. 4

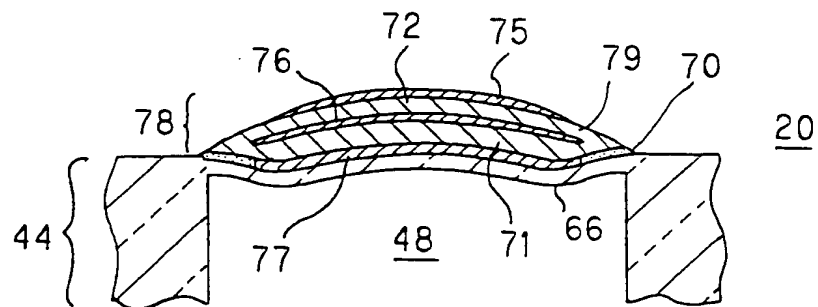




FIG. 5

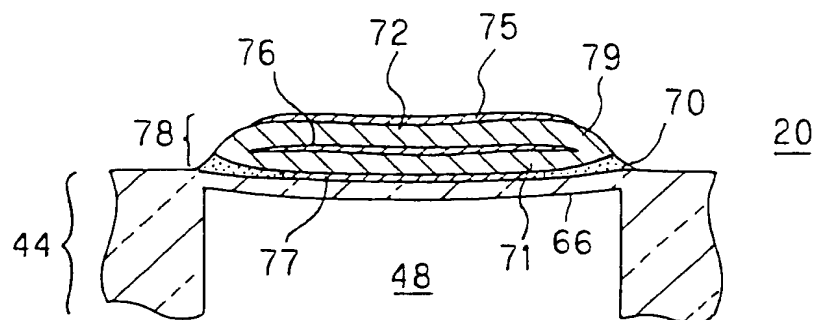


FIG. 6

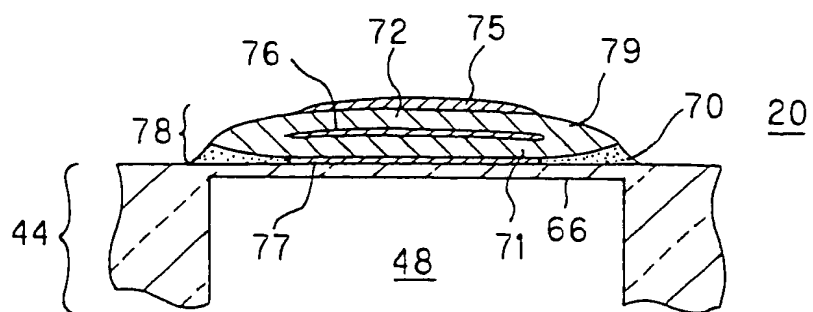


FIG. 7

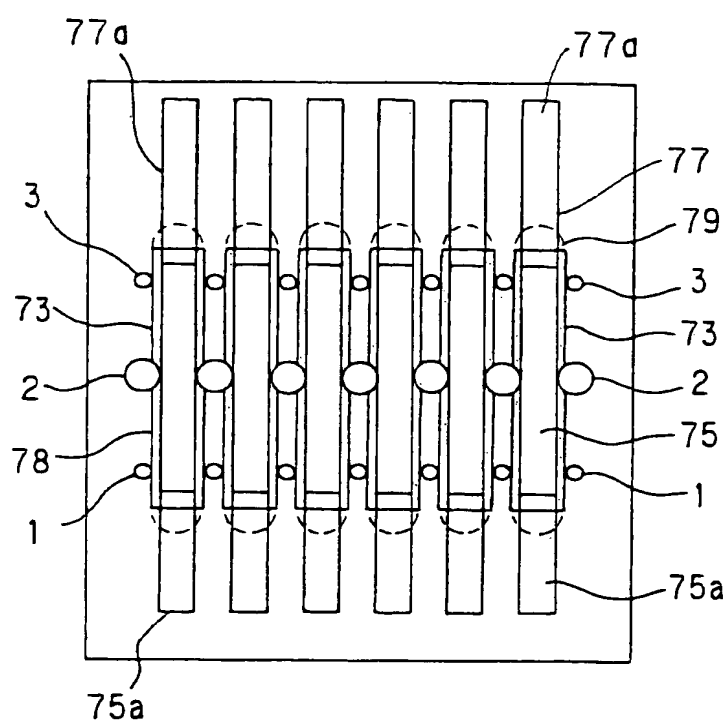


FIG. 8(a)

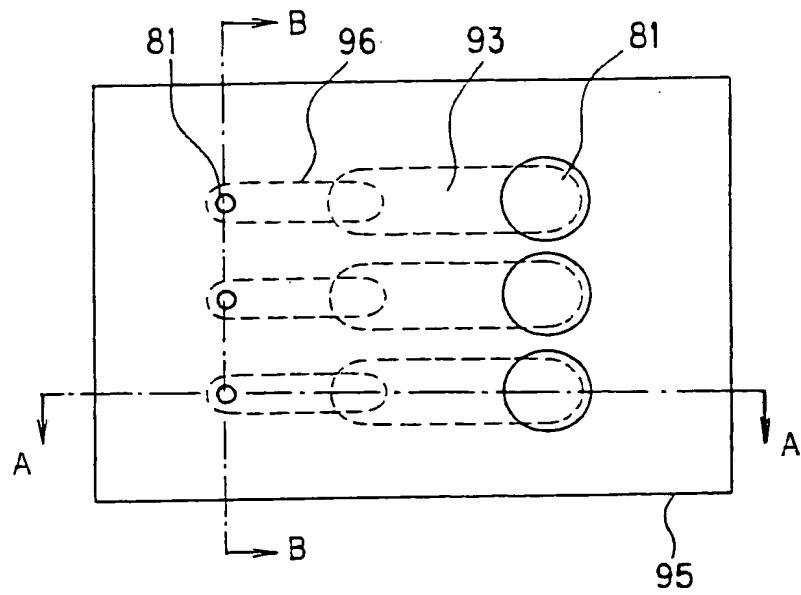


FIG. 8(b)

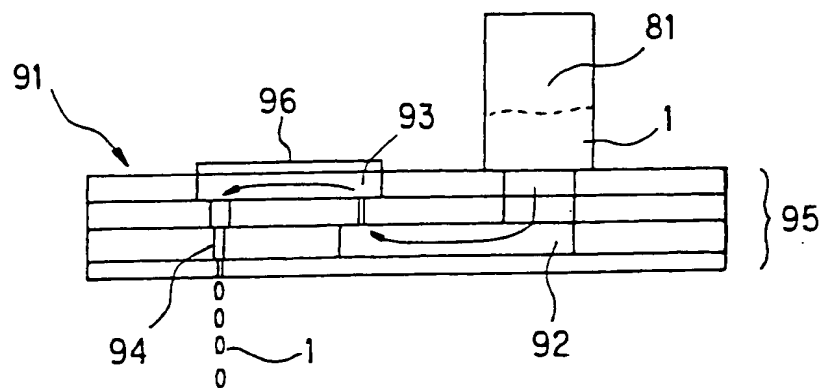


FIG. 9(a)

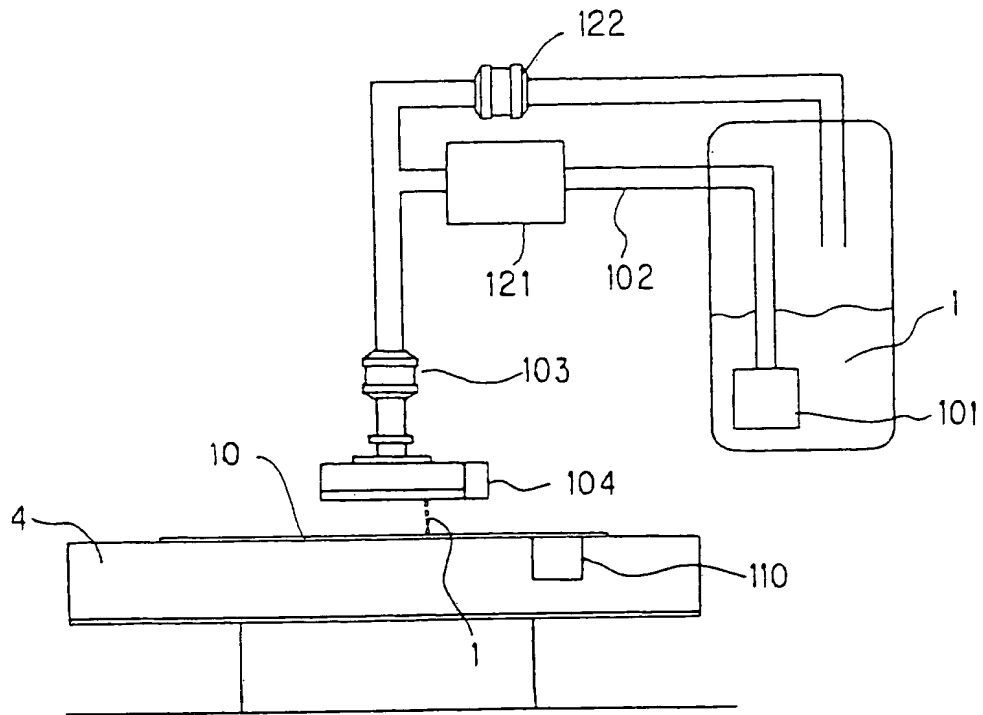


FIG. 9(b)

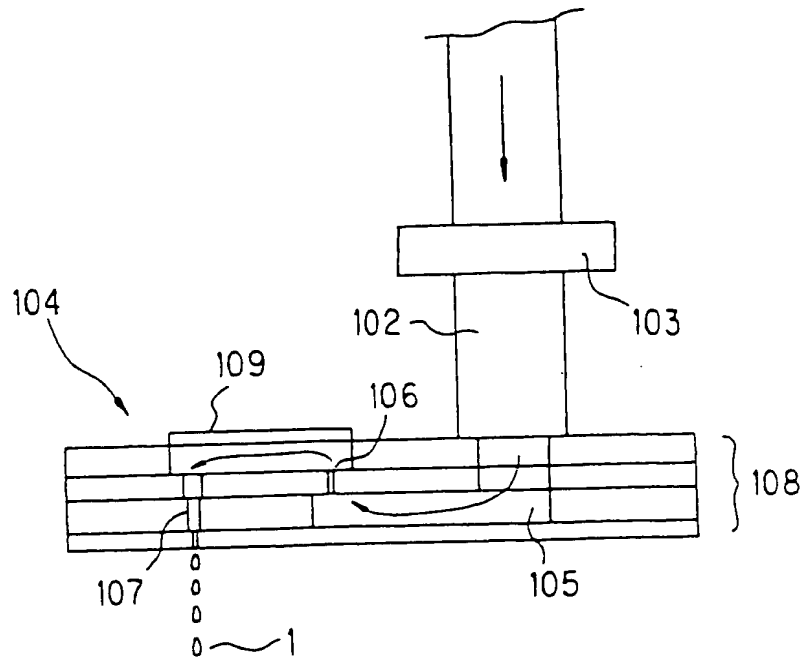


FIG. 10

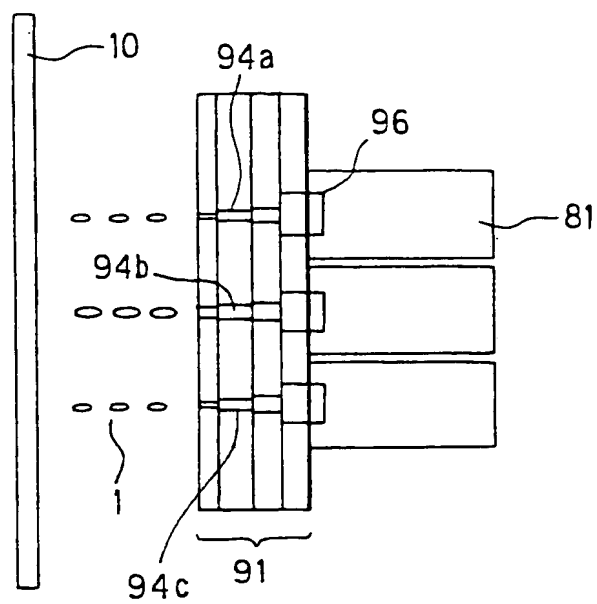


FIG. 11

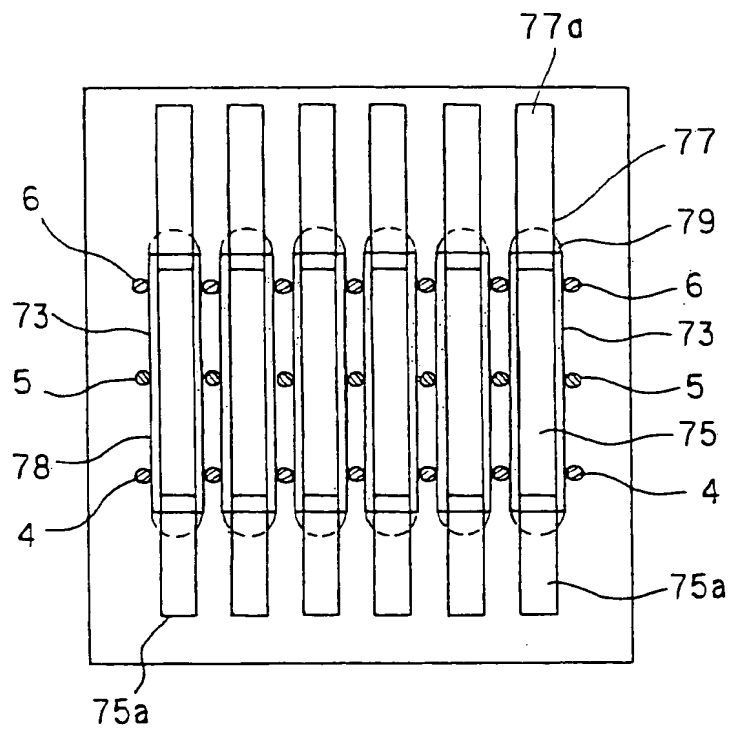


FIG. 12

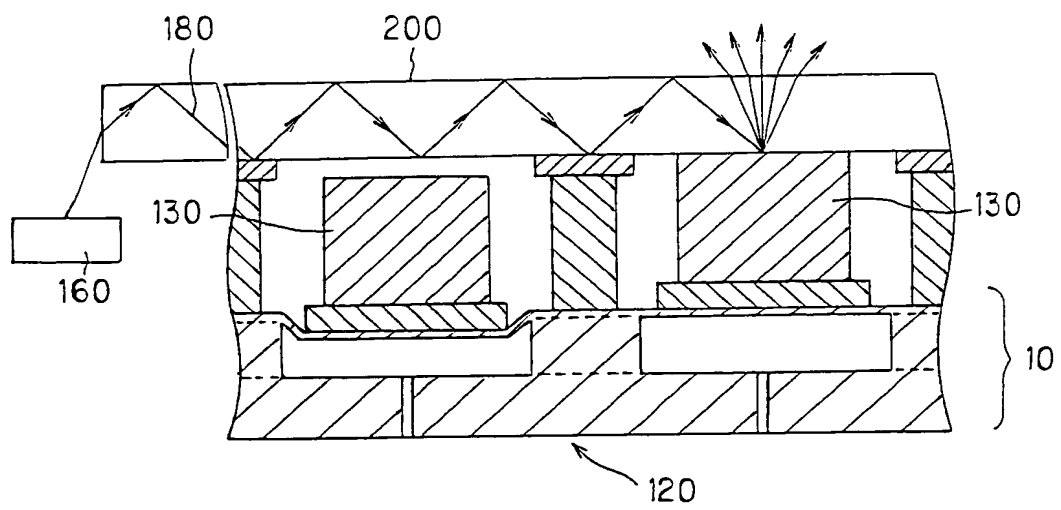


FIG. 13

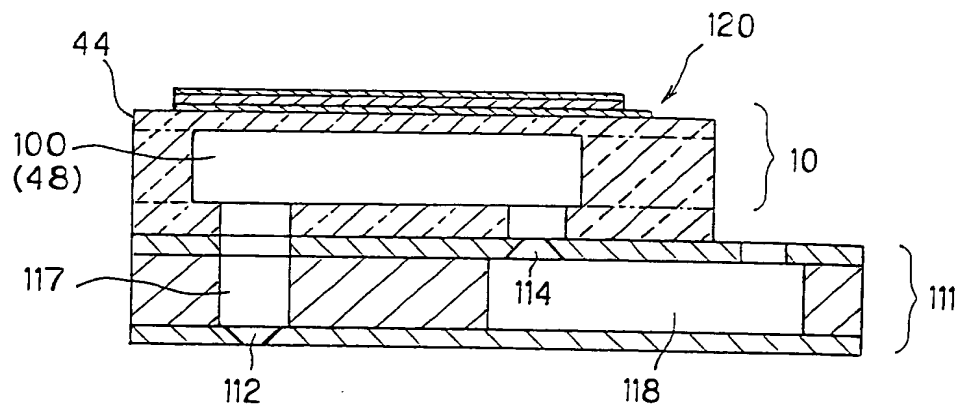




FIG. 14(a)

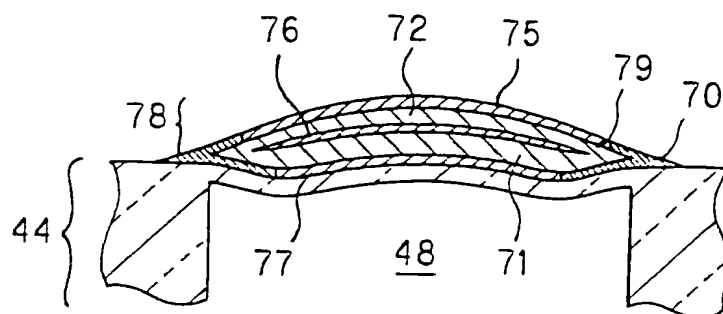


FIG. 14(b)

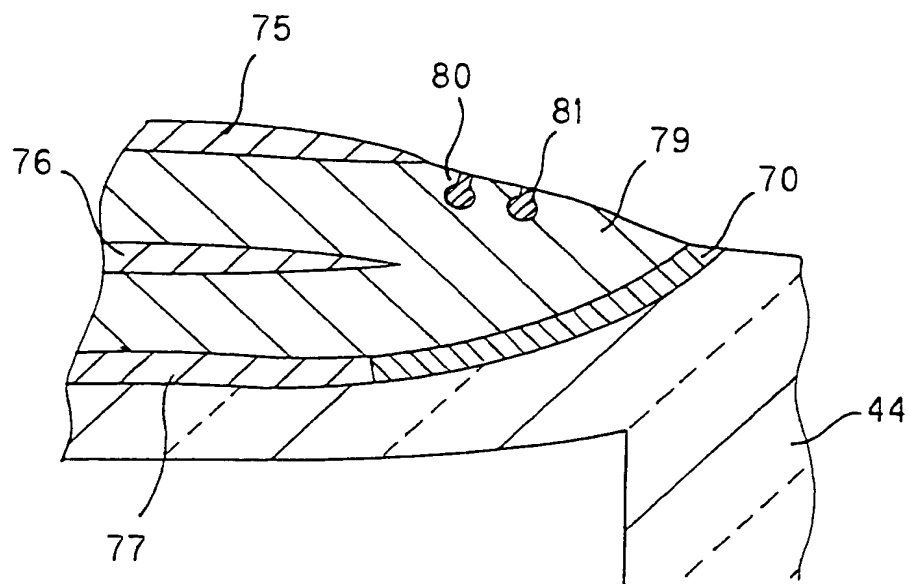


FIG. 15

